CRUMB RUBBER MODIFIED BITUMEN (CRMB): INFLUENCE OF THE SELECTION OF RAW MATERIALS & RHEOLOGICAL PROPERTIES

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ABSTRACT

The use of crumb rubber from old tyres in the production of asphalt mixes has become a very interesting option for the modification of bitumen in order to obtain asphalt mixes of higher quality and, at the same time, solve an environmental problem.
Spain disposes of an extensive legal framework regarding the specifications for this type of binders and the mixes prepared with crumb rubber, as well as some recommendations during production and storage of these new binders.
Not only the nature of the raw materials used for the development of these new binders, but also the process conditions for their production, has turned out to be of great importance. The influence of the crumb rubber (composition, grading, density, manufacturing process,…) and the asphalt bitumen used has to be taken into account, because although it is possible to comply with the specifications defined for this type of binders, there is no guarantee of compatibility among their components.
This communication presents the results obtained from the empirical and rheological characterization of different crumb rubber modified binders (BC and BMC) produced with different raw materials to evaluate their behavior in the asphalt mixes (cohesion, ageing, rutting resistance and fatigue).
The main goal is to know as much as possible about these binders and see the influence that the raw materials have in their final characteristics.
The aim of this communication is that these binders become of normal use in the different worksites, leaving behind the experimental character that their use has nowadays.

Keywords: crumb rubber, rheological properties, compatibility, rutting resistance

INFLUENCE OF THE CHARACTERISTICS OF THE CRUMB RUBBER ON THE QUALITY OF CRUMB RUBBER MODIFIED BITUMEN (CRmB)

1. INTRODUCTION

The capacity for the crumb rubber (CR) from old tyres to improve the properties of the bituminous binders is based on the interaction between the crumb rubber particles and bitumen. The characteristics of these raw materials are vital. There are other process conditions, such as temperature or reaction time, which are to be taken into account as well.
This article shows the properties of different products obtained from the same base bitumen and two types of CR.
Both types of CR come from a mechanical crushing process, but there is a big difference between them: their composition. While one of them is 100% truck tyre, the other is a mix of unknown proportions of truck and car tyre.
They also have different grading. The smaller the particles, the better and quicker they *react* with bitumen. The tiniest CR particles swell when they absorb bitumen oils, by becoming softer and then bigger. Viscosity increases and stabilizes at the end of the production process, so that the final product does not settle during storage.
According to the CR composition, truck tyres have a bigger proportion of natural rubber than car tyres. Natural rubber is more compatible with bitumen than synthetic ones (SBR, BR, IR, etc.). From now on, **CR 100% from truck tyres** will be known as *compatible CR* and **crumb rubber from truck and car tyres** will be known as *non-compatible CR*.
The main characteristics of these raw materials are shown in Table 1:

Table 1.Characteristics of crumb rubber

Crumb Rubber	Compatible	Non compatible
Maximum size (mm)	0,5	0,8
Composition (%)	100% truck tyre	Unknown % of truck and car tyre
Natural rubber content (%)	36	30
Textile content (%)	< 0,2	< 0,5

Several CR modified binders (BC and BMC) have been produced with these raw materials and different formulations, showing different properties from the point of view of storage stability.
The main goal of this study is to prove the different behavior of these binders, depending on the type of CR used, and take this fact into account to preview a difference in behavior of the asphalt mixes prepared with them, especially regarding rutting resistance, fatigue and thermal cracking.

2. STUDIED BINDERS

The binders studied for this article are the following ones:

- *Non compatible* CR modified binders: BC 35/50, BC 50/70 and BMC-2.
- *Compatible* CR modified binders: BC 35/50 BC 50/70 and BMC-2.
- Base bitume: B 35/50 and B 50/70 (reference).

Only the results of BC 35/50 are going to be presented, as there is no difference with the ones obtained for BC 50/70 or BMC-2 for the purpose of this study.

3. EXPERIMENTAL DEVELOPMENT

Apart from the empirical characterization of the fresh binder, the following test procedures have been used:

3.1 Ageing process

All the binders have undergone several ageing processes:

- Resistance to hardening under the influence of air and heat– RTFOT (EN 12607-1), to simulate the ageing of the binder during the mix production in the plant.
- Accelerated long-term ageing conditioning by a Pressure Ageing Vessel – PAV (EN 14769), to simulate the ageing of the binder after several years on the road. These binders come from the previous ageing process.

3.2 Rheological characterization

3.2.1 Using a Dynamic Shear Rheometer **(DSR),** according to the *EN 14770*standard– *Determination of complex shear modulus and phase angle. Dynamic Shear Rheometer (DSR).*

This test method allows the evaluation of the viscoelastic behavior of bituminous binders using the complex modulus, *G**, as the relation between the stress applied to the binder and the strain suffered by it, and the phase angle, δ, which gives an idea of the recoverable and non-recoverable strain of the binder.

These parameters are measured by the rheometer's software when applying an oscillating movement to the sample, "sandwiched" between parallel plates (one fixed, the other that oscillates) with a specific thickness, at different frequencies and test temperatures. The sample temperature keeps constant thanks to a water bath which surrounds the sample during the entire test.

All the samples have been tested using a frequency sweep (0,10, 0,16, 0,25, 0,40, 0,63, 1,00, 1,59, 2,51, 4,00, 6,32 y 10,00 Hz) at different test temperatures, divided in two groups:

- The first sweep has been carried out at intermediate service temperatures (10, 20, 30 and 40ºC) with the 8mm parallel plate and a sample thickness of 2mm.
- The second sweep has been carried out at high service temperatures (40, 50, 60, 70 and 80ºC) with the 25mm parallel plate and a sample thickness of 1mm.

The results are represented using:

- *Black Diagram*: variation of the magnitude of the complex modulus, G*, versus the phase angle, δ.
- *Isochronal variation of G* and* δ as a function of temperature, at a constant frequency of 1,59 Hz (equivalent to cars passing over the road at a speed of 90 Km/h).
- *Fatigue cracking*: isochronal variation of $G^*\sin\delta$ as a function of temperature, at a constant frequency of 1,59 Hz (equivalent to cars passing over the road at a speed of 90 Km/h).

3.2.2 Using a Bending Beam Rheometer (BBR), according to the *EN 14771 standard – Determination of the flexural creep stiffness. Bending Beam Rheometer (BBR).*

This test method allows the measurement of the stiffness of the binder at low service temperatures using the stiffness modulus, *S*, as the relation between the stress applied to the binder and the flexural creep stiffness suffered by it, and the m-value, as the variation of the stiffness of the binder during the application of the flexural stress.

These parameters are measured by the rheometer's software when applying a constant load at the midpoint of the test sample, which is supported at two points, during a specific period of time at a selected temperature.

The sample temperature keeps constant thanks to a liquid bath which surrounds the sample during the entire test.

3.3 Dynamic viscosity measurements

According to the EN 13302 standard. It gives information about the relative resistance presented by the spindle to rotate inside a cylinder containing a certain quantity of sample at a specific test temperature.

4. PRESENTATION OF RESULTS

After the empirical characterization of the fresh binders mentioned before (compatible and non-compatible BC 35/50), their characteristics have been compared with the ones from a base bitumen with the same penetration grade (B 35/50). There is also a comparison of the rheological properties of the binders before and after ageing (RTFOT and PAV) using the Black diagram and the isochrones of G*, δ and G*sinδ VS temperature:

- ***Black diagram:*** there are two distinctive behaviors between a conventional bitumen and a CR improved/modified one (BC/BMC). While a conventional binder shows a lineal tendency, the modified ones have a characteristic *S*-shape, due to the interaction of the elasticity of the elastomeric polymer on the viscoelastic behavior of the modified binder. For the same G* value, δ values are lower for these binders – higher rheological elasticity.
- This tendency is also reflected in the isochrone of δ at 1,59 Hz, where this value increases lineally with temperature for a conventional binder (higher viscosity). In the case of modified binders, this curve keeps the *S*-shape.

  In relation to the complex modulus values at 1,59 Hz, this tendency is always lineal for any type of binder, modified or not, always increasing with the decrease in temperature. This time, the difference among binders resides in the gradient of the curve. The lower the gradient, the better the thermal behavior of the binder is going to be.

  The isochrone of **G*sinδ** is lineal and decreasing with the increase in temperature. It gives the temperature at which the aged binders after RTFOT and PAV comply with the SHRP specification for fatigue cracking: G*sinδ=5000 kPa
- Determination of the PG grade of the studied binders, according to the SHRP specification, which considers the following service temperatures:
  - Temperature at G*/sinδ=1KPa for the fresh binder.

- Temperature at S=300MPa and m=0,3 for the aged binder after RTFOT and PAV.

5. FINAL RESULTS

5.1 BC 35/50

- *Fresh binder*

According to the data shown in table 2, the two types of BC 35/50 have a similar penetration value compared to the one of the base bitumen taken as a reference, but the R&B values are clearly higher, giving a better penetration index as a result. Both binders also comply with the specification in relation to the storage stability, although the non-compatible (NC) BC 35/50 presents a tendency to phase separation during storage if there is not a previous homogenization by stirring.

The viscosity for both CR modified binders is higher than that of the base bitumen, due to the presence of CR from old tyres.

Table 2. Empirical characteristics of the binders

CHARACTERISTICS	UNIT	TEST METHOD	Compatible BC 35/50	Non-compatible BC 35/50	B 35/50	Specs BC 35/50
Fresh boinder:						
* Penetration (25ºC; 100 g; 5 s)	0,1 mm	EN 1426	41	43	41	35-50
* Softening point (R&B)	ºC	EN 1427	60,2	60,8	54,6	≥ 58
* Penetration index	-	EN 12591	0,6	0,9	-0,5	
* Storage stability		EN 13399				
- Difference in R&B	ºC	EN 1427	4	5,4	-	≤ 10
- Difference in Penetration (25ºC)	0,1 mm	EN 1426	7	5	-	≤ 8
* Elastic recovery (25ºC; torsion)	%	EN 13398	34	21	-	≥ 10
* Dynamic viscosity	mPa.s	EN 13302				
@ 135 ºC			1415	1625	700	
@ 150 ºC			650	695	250	
@ 175 ºC			210	255	100	

The Black diagram (figure 1) shows the lineal tendency for the base bitumen B 35/50, while the two BC 35/50 have this S-shaped curve more proper of a polymer modified binder, with lower δ values for the same G* values.

The NC BC 35/50 has a similar behavior than the compatible BC 35/50. These results are justified, because although there is a phase separation in the sample on the NC BC 35/50 (high amount of CR concentrated at the bottom), there is a need to homogenize the sample to run the tests, so that both phases (bitumen above and CR below) mix again.

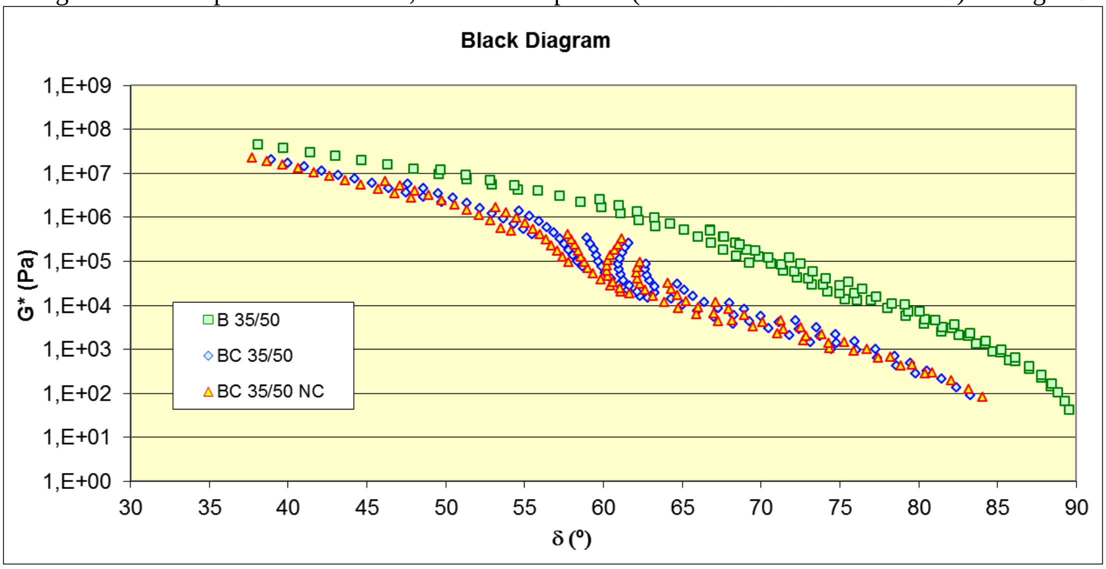

Figure 1. Black diagram – fresh binder

The isochronal variation of G* and δ VS temperature (figure 2) shows how the viscoelastic properties of both BC 35/50 are better than the ones of the reference bitumen, mainly the δ values, which are lower for all the test temperatures (more elastic binders).

The thermal susceptibility for the CR modified binders improves slightly with the presence of crumb rubber; at intermediate service temperatures, G* values are lower than the ones showed by the base bitumen and higher at high service temperatures. This means a higher stiffness and a better behavior of the CR modified binder on the road.

Like in Figure 1, there is not a noticeable difference in the viscoelastic behavior between the compatible an NC BC 35/50, due to the necessary homogenization of the samples to run the tests.

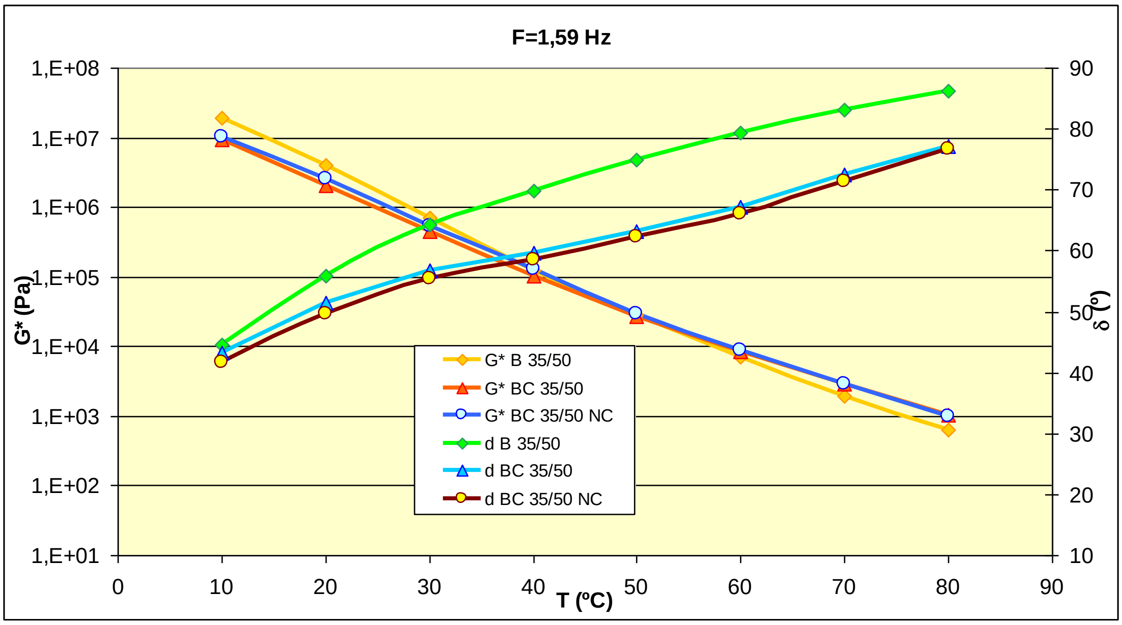

Figure 2. Isochrones of G* and δ - fresh binder

- *Aged binder*

After the ageing process, there are bigger differences between the compatible and NC BC 35/50 (figures 3 and 4).

The NC BC 35/50 shows lower δ values (more elastic behavior), while the complex modulus is higher at all service temperatures than for the rest of binders after PAV ageing.

The fact that the NC BC 35/50 is more elastic than the compatible BC 35/50 may be caused by the presence of a higher content of CR from car tyre used to produce this CR modified binder. Truck tyres have a bigger percentage of natural rubber, which is less elastic than the synthetic rubber present in car tyres.

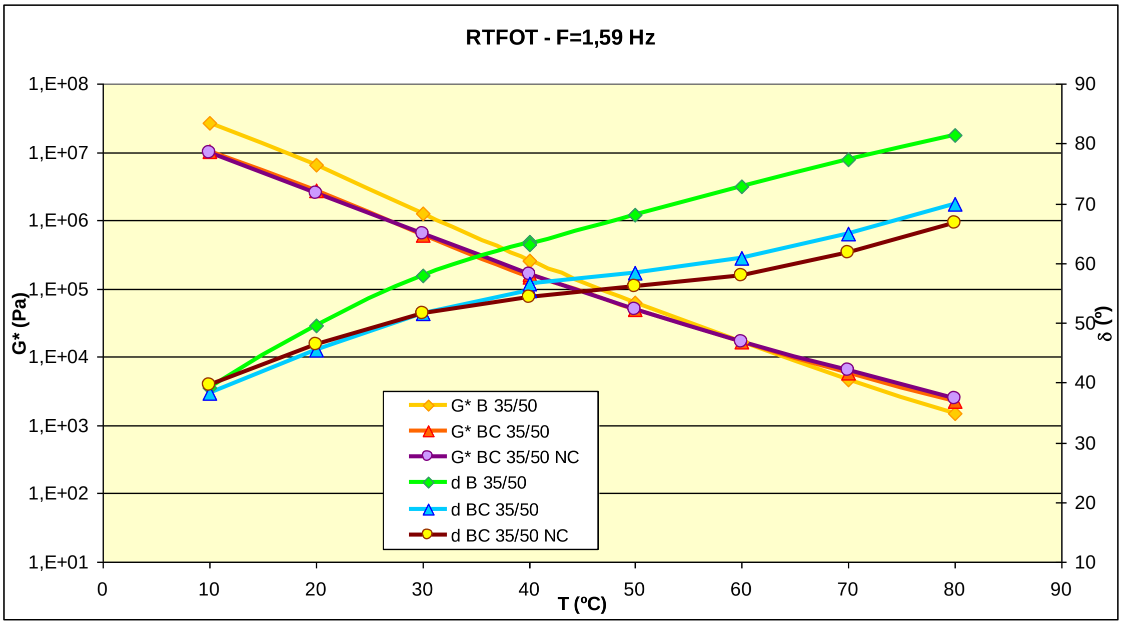

Figure 3. Isochrones of G* and δ - RTFOT aged binder

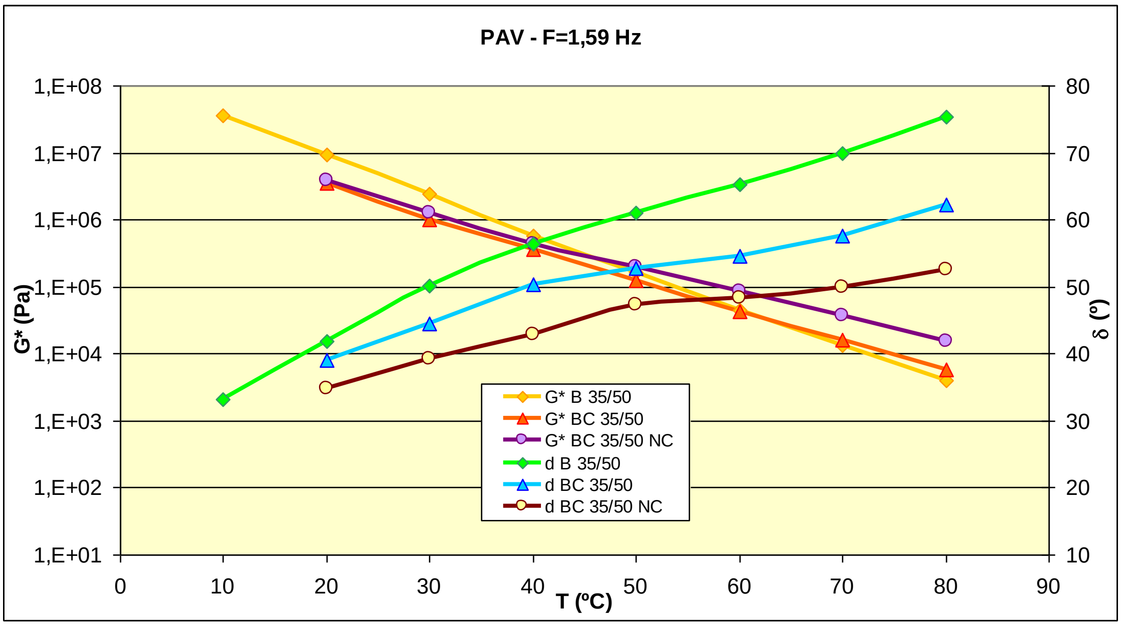

Figure 4. Isochrones of G* and δ - RTFOT & PAV aged binder

Fatigue cracking

Figure 5 reflects the temperature at which the different BC 35/50 are going to start presenting fatigue cracking resistance, due to the continuous bending of the pavement layers caused by the traffic. These values are lower than the one measured for the bitumen taken as a reference (table 3). The rheological properties for the CR modified binders are better than the ones shown by the base bitumen with a similar penetration.

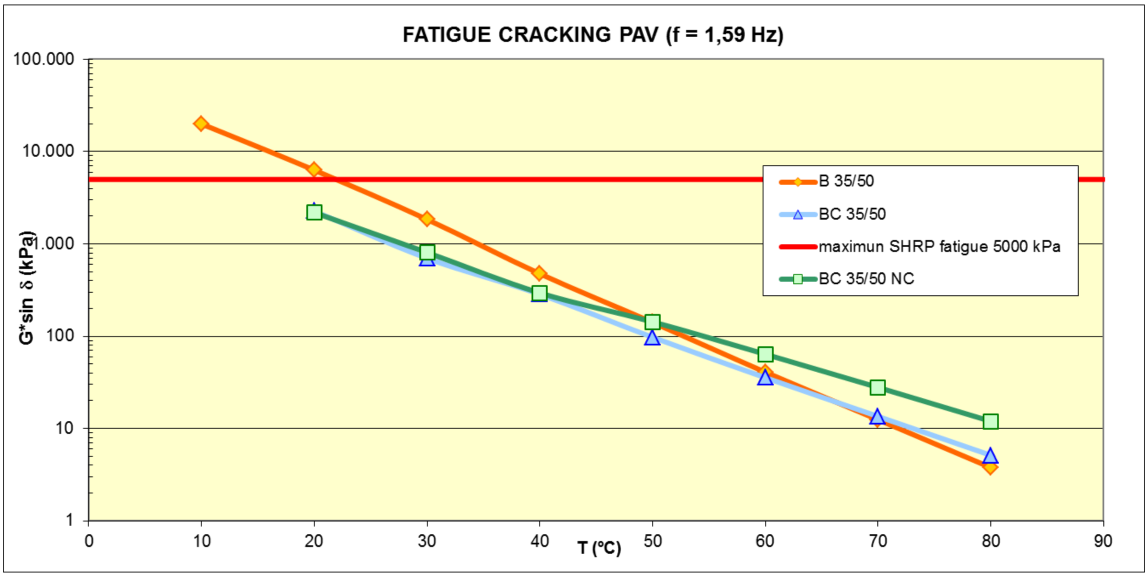

Figure 5. Fatigue cracking – RTFOT & PAV binder

Table 3. Minimum fatigue cracking temperature

	B 35/50	Compatible BC 35/50	Non compatible BC 35/50
Temperature @ G*sinδ=5000 kPa (ºC)	21,4	11,3	8,8

Thermal cracking
Cracks may appear on the pavement at low service temperatures, due to the contraction of the different layers which conform it. According to the SHRP specification, a maximum stiffness of S = 300 MPa and a minimum relaxing gradient of m = 0,3, both measurements taken after 60s of load application, are considered. The temperatures that comply with this double condition are shown in Table 4:

Table 4. Minimum thermal cracking temperature

	B 35/50	Compatible BC 35/50	Non compatible BC 35/50
Temperature @ S=300 Mpa & m=0,3 (ºC)	-10	-16	-14

The complete rheological characterization gives the following PG grades for the studied binders:

Table 5. PG grades

	B 35/50	Compatible BC 35/50	Non compatible BC 35/50
PG GRADE	70-16	76-22	76-22

There are no significant differences between the two CR modified binders, in spite of the non-compatibility of one of them.

5.2. RESULTS OF A NON-COMPATIBLE BC 35/50 “WITHOUT PREVIOUS HOMOGENEIZATION”

After not having found any outstanding difference between the compatible and NC BC 35/50 from the rheological point of view, new tests have been run on a NC BC 35/50, but this time the sample preparation methodology has changed. The target is to reproduce what we may find inside a storage tank: the quality of the binder in the upper part and at the bottom. Storage stability samples have been cut in half and the different parts have been tested individually to measure their empirical and rheological properties (these samples have been processed after a substantially longer time than the one specified in EN 13399 to reproduce what can really be found inside a storage tank on the worksite).

- *Fresh binder*

As it is shown in table 6, the binder from the upper part of the tank is mainly bitumen with a small portion of CR, so that its behavior is closer to the one shown by the base bitumen with the same penetration grade. The softening point, as well as viscosity, is a bit higher, due to the presence of this little percentage of CR.
The bottom part contains a much higher percentage of CR, settled during storage. The penetration of this fraction is very close to the upper limit of the specification, because the CR absorbs the maltenes from the bitumen. The softening point and the elastic recovery are also very high, due to this greater amount of CR present in the sample. Viscosity is very similar as the one which would present a CR highly modified binder (BMC-3c).

Table 6. Empirical characteristics of the binders

CHARACTERISTICS	UNIT	TEST METHOD	Compatible BC 35/50	Bottom	Top	B 35/50
Fresh boinder:						
* Penetration (25ºC; 100 g; 5 s)	0,1 mm	EN 1426	41	49	45	41
* Softening point (R&B)	ºC	EN 1427	60,2	64,2	59,2	54,6
* Penetration index	-	EN 12591	0,6	1,5	0,6	-0,5
* Elastic recovery (25ºC; torsion)	%	EN 13398	34	42	25	-
* Dynamic viscosity	mPa.s	EN 13302				
@ 135 ºC			1415	7100	885	700
@ 150 ºC			650	4000	445	250
@ 175 ºC			210	1510	150	100

Figure 6 shows the differentiated behavior of the fractions. The upper fraction shares a lineal tendency with a base bitumen, but with a slight S-shape thanks to the small percentage of CR present in it.
On the other hand, the bottom fraction has a better marked S-shaped curve, typical of a highly CR modified binder, given the quantity of CR contained in it, and very close to the one shown by a highly polymer modified binder (BM-3c), although this last binder has a slightly better elastic response.

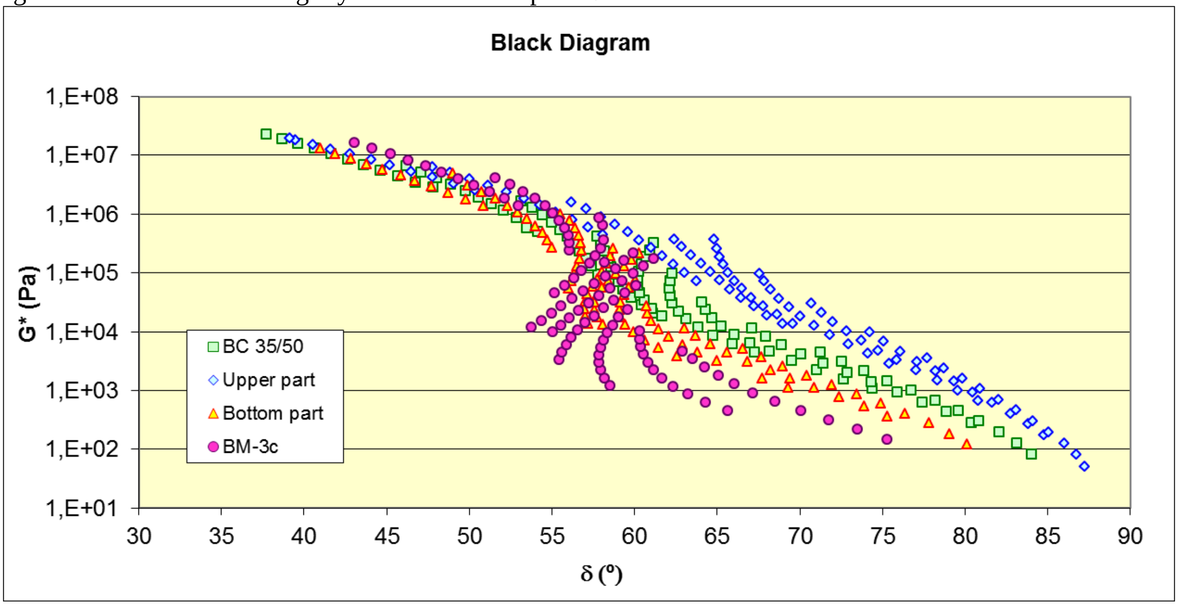

Figure 6. Black Diagram – fresh binder

The isochrone (figure 7) let us see the intermediate behavior of the compatible BC 35/50 between the two fractions very clearly. G* values are very similar for the three binders, just with a small difference in the curve gradient: better thermal behavior for the binder at the bottom of the storage tank.
In relation to δ values, the binder with the best rheological behavior is the one from the bottom fraction, due to the higher concentration of CR (very elastic binder).

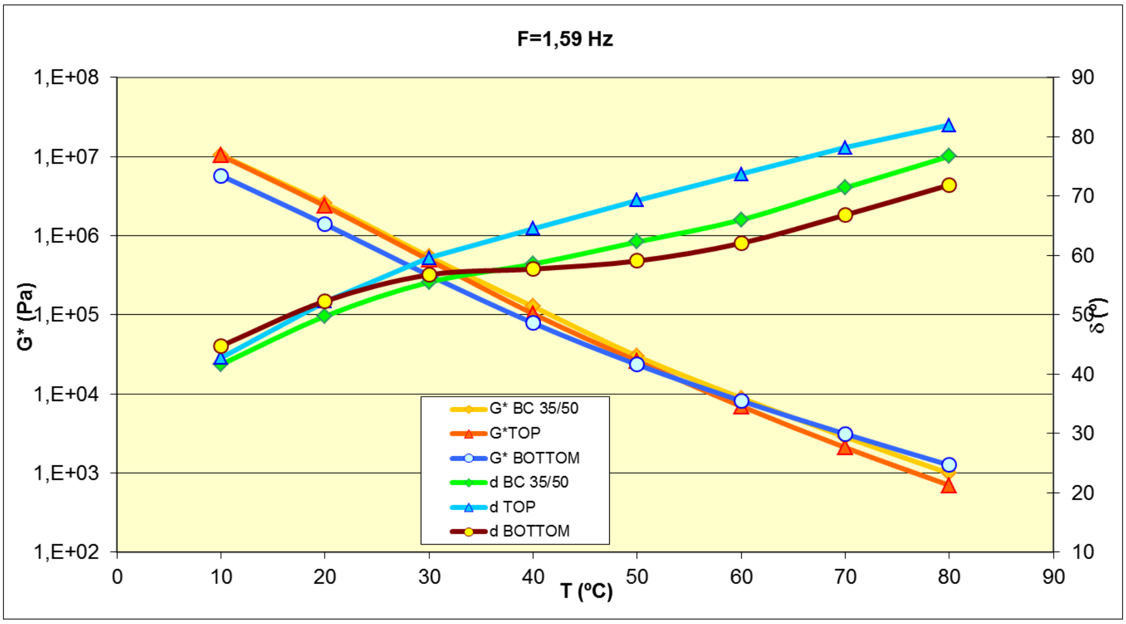

Figure 7. Isochrones of G* and δ - fresh binder

➢ *Aged binder*

After completing the rheological characterization of all the binders using the DSR and the BBR, the final PG grades for each of them are the following ones:

Table 7. PG grades

	Compatible BC 35/50	Bottom	BMC-3c*	Top	B 35/50
PG GRADE	76-22	82-22	82-28	76-16	70-16

(*) Highly CR modified bitumen

The amount of CR in the bottom fraction is so elevated, that its rheological behavior can be compared with the one presented by a CR highly modified binder, type BMC-3c.

6. CONCLUSIONS

According to all the data gathered throughout this study, we can conclude saying that:

- Although all the binders comply with the specifications for this type of products, there are existing differences in relation to the storage stability.
- CR modified binders which using CR from truck tyres present better storage stability, thanks to the presence of a higher percentage of natural rubber.
- There are no significant differences in the rheological behavior of compatible and NC CR modified binders, because of the necessity to homogenize the sample to run the tests. This operation hides any possible way to distinguish both types of binders.
- The final PG grade is the same for each binder, independently of their compatibility, when the tests are run on previously homogenized samples.
- For NC CR modified binders, there is a different rheological behavior of the binder, depending on where the sample has been taken from: the upper part of the storage tank or the bottom. The upper fraction is similar to a base bitumen with the same penetration grade. Meanwhile, the lower fraction has a very high viscosity, comparable to the one of a CR highly modified binder, type BMC-3c. This would mean the use of powerful pumping devices that were able to move this product.
- It is, therefore, necessary to count on an adequate stirring system to prevent a possible phase separation on this type of binders, in order to guarantee a homogeneous quality of the supplied product, apart from avoiding problems of residues at the bottom of the storage tanks.
- There is one more relevant thing to say about crumb rubber: the particle size. For the same mechanical crushing process, the smaller the particle size is, the greater the specific surface is, so that it reacts faster and better with bitumen. Crumb rubber, mainly from truck tyres and with a very small particle size, improves the compatibility of the final product.

To finish with, it is fair to say that it is possible to prepare asphalt mixes using CR modified binders with good mechanical properties, even if non compatible products are being used. In that case, it is necessary to count on stirring or recirculating systems, which avoid the CR settling down at the bottom of the storage tank.
Not taking these advices would mean the risk to supply asphalt mixes with non-homogeneous quality.

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